

Declaration

- I, Kenji Tokuhisaha, as one of the inventors of the subject invention, declares that I made a statement regarding the reasons for rejection by the examiner in charge.
- 1. The method for producing fluorine-containing saturated aliphatic carboxylic acids in accordance with Fuchikami patent cannot produce any alkoxy fluorine-containing propionic acid ester. This is clear from the following experimental example.

 EXAMPLE

$$C_4H_9$$
 C_8F_{17}
 I
 C_4H_9
 C_8F_{17}
 C_8F_{17}
 $COOE$

An autoclave was charged with $(Ph_3P)_2PdCl_2$ (34.8 mg, 0.05 mmol), $C_8F_{17}CH_2CH(C_4H_9)I$ (360 µL, 1 mmol), Et_3N (0.14 mL, 1.0 mmol), and EtOH (1 mL). The system was pressurized with carbon monoxide to 3 MPa, and the reaction mixture was then stirred at 80°C for 12 hours. The reaction mixture was purified by silica gel column chromatography and ethyl 2-butyl-3-perfluorooctylpropionate was isolated (0.43 g, 75% yield). However, no alkoxy fluorine-containing propionic acid ester was yielded. This experimental example is cited from the description in EXAMPLE 10 of column 8 of the Fuchikami patent.

2. The method for producing fluorine-containing acrylic acid esters in accordance with Matteoli article produces as a main product an alkoxy fluorine-containing propionic acid ester whereas the method in accordance with the present invention can produce only a small amount of alkoxy fluorine-containing propionic acid ester and as a main product fluorine-containing acrylic acid ester. This can be proved clearly by the following experimental example.

EXAMPLE

$$= \begin{array}{c} CF_3 \\ Br \end{array} + CO + ROH \longrightarrow \begin{array}{c} CF_3 \\ COOR \end{array} + \begin{array}{c} CF_3 \\ COOR \end{array}$$

An autoclave was charged with CH2=C(CF3)Br (5.7 mmol), (Ph3P)2PdCl2 (0.1 mmol), an alcohol (7.2 mmol), and a base (Bu_3N or Pr_3N , 6.3 mmol). The system was pressurized with carbon monoxide to 5 MPa, and the reaction mixture was then stirred at 100°C for a predetermined period of time. I analyzed the reaction mixture to determine the products listed in the following table as Entries 1 to 5. These experiments were made in accordance with the production method of Matteoli article to produce fluorine-containing acrylic acid esters, and this example is cited from the description in Table 4 on page 289 of Matteoli article. In the same manner, an autoclave was charged with $CH_2=C(CF_3)Br$ (1.0 mmol), $(Ph_3P)_2PdCl_2$ (0.01 mmol), an alcohol (1.2 mmol), basees (Et₃N 1.0 mmol) + Li_2CO_3 0.1 mmol), and THF (2 mL). The system was pressurized with carbon monoxide to 1 MPa, and the reaction mixture was then stirred at 100°C for 5 hours. The reaction mixture was analyzed and the product was determined and listed in the following table as Entry 6. This example was made in accordance with the production method of the present invention to produce fluorine-containing acrylic acid esters.

Entry	Base	Alcohol	Time h	Conversion %	Yield	
					1 (%)	2 (%)
1	Bu ₃ N	Methanol	15	84	24	49
2	Bu ₃ N	2-Propanol	66	96	36	32
3	Bu ₃ N	1-Butanol	3	98	20	40
4	Bu ₃ N	2-Butanol	6	93	16	38
5	Pr ₃ N	2,2-Dimethylpropanol	15	71	17	40
6	Et ₃ N+Li ₂ CO ₃	Ethanol	5	100	82	3

As is clear from the results in the above table, the combined use of two types of bases like the present invention can drastically change the production ratio between the two types of products, thereby significantly improving the yield of the target product, fluorine-containing acrylic acid esters when compared to the yield obtained by the conventional methods.

As discussed above, the present invention employs the combination of arbitrary bases to drastically improve the yield of

fluorine-containing acrylic acid esters, and this cannot be expected by the teachings of the cited references.

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